National Analytical Management Program (NAMP)
U.S. Department of Energy Carlsbad Field Office

Radiochemistry Webinars

Environmental/Bioassay Radiochemistry Series

Gamma Spectrometry (Part 2)

In Cooperation with our University Partners

Meet the Presenters…

Robert Litman

Robert Litman, Ph.D., has been a researcher and practitioner of nuclear and radiochemical analysis for the past 42 years. He is well respected in the nuclear power industry as a specialist in radiochemistry, radiochemical instrumentation and plant systems corrosion. He has co-authored two chapters of MARLAP, and is currently one of a team of EMS consultants developing radiological laboratory guidance on radionuclide sample analyses in various matrices, radioactive sample screening, method validation, core radioanalytical laboratory operations, contamination, and rapid radioanalytical methods. He authored the section of the EPRI PWR, Primary Water Chemistry Guidelines on Radionuclides, and has been a significant contributor to EPRI Primary-to-Secondary Leak Detection Guidelines. Dr. Litman has worked with the NRC in support of resolving GSI-191 issues (chemical effects following a loss of coolant accident) at current nuclear power plants and reviewed designs for addressing that safety issue for new nuclear power plants. His areas of technical expertise are gamma spectroscopy and radiochemical separations. Dr. Litman has been teaching courses in Radiochemistry and related special areas for the past 28 years.

Bob Shannon

Mr. Bob Shannon has supported government and independent commercial testing laboratory radiochemistry needs for over 20 years and currently performs consulting work through Environmental Management Support (EMS) and his company, QRS, LLC. His recent project support includes drafting revision 2 of NRC RG 4.15 to incorporate MARLAP principals, developing and teaching training courses on basic radiochemistry for State and Federal lab radiochemists, performing audits for the EPA and DOE, and helping author laboratory guidance documents and develop Rapid Radioanalytical Methods for the EPA. Mr. Shannon chairs The NELAC Institute *Radiochemistry Expert Committee*, the ASTM D19.04 *Fission and Activation Products Task Group*, and is the Radiochemistry Part Coordinator for *Standard Methods for the Examination of Water and Wastewater*.

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Gamma Spectrometry Part 2

Analytical Libraries,
Spectral Review and Interpretation,
Random and Coincidence Summing

Robert Litman, PhD
Bob Shannon

EMS

National Analytical Management Program (NAMP)
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TRAINING AND EDUCATION SUBCOMMITTEE

Gamma Spectrometry Part 2

This webinar is based on a workshop presented at the 2012 RRMC by Bob Shannon, Doug Van Cleef, Dave Burns and Bob Litman

Part 2 Topics

- Libraries
  - Nuclear Constants
  - Software Functions
  - Examples
- Activity Calculations and Spectrum Review
- Summing in Gamma Spectrometry

Libraries

Nuclear Constants, Software Functions, and Examples

Gamma Spectrometry Library

- Collection of nuclear constants
  - Half-life
  - Gamma ray energies*
  - Abundance for each gamma ray energy listed

- One main library initially, user can create additional ones based on need

**Not all gamma ray energies for all listed radionuclides are included*

Selection of Nuclides and Lines for Sub-Libraries

- From the main library, create sub-libraries
  - Sample or client specific
    - Radionuclides known for that sample?
    - Radionuclides to be reported even if not detectable?
    - Which gamma rays for qualification/quantification?
    - Gamma rays used for quantification within calibration curve?
    - Interference correction needed?

Gamma Spectrometry Libraries (cont.)

- User-defined adjustments to nuclear constants are often needed to:
  - Adjust abundance to reflect branching ratios
  - Adjust half-lives to reflect known (or assumed) radioactive equilibrium for a matrix or sample type
  - Adjustments to reflect inferred analytical assumptions; or
  - Various combinations of these
- Project-specific library entries may require further changes to accommodate special circumstances

Other Library Information

- Dependent upon software and analyst selection
  - MDA for each radionuclide
  - Use key line or weighted average
  - Fractional abundance limit
  - Tentative ID of “unknown” gammas may revert to the main library or a specific secondary library

Minimum Detectable Activity
Minimum Detectable Concentration

- Both concepts are *a priori* determinations
  - MDA is an activity: Bq or pCi
  - MDC is a concentration: Bq/L or pCi/L
  - Identifies *potential* instrument/method capability
  - Uses nominal parameters
- ***The MDA/MDC is not to be used to make detection decisions!!!***

Radionuclide Half-Life

- Used by software algorithm to calculate sample activity based on peak area, count time, efficiency , *decay correct to sample count start*, and *decay correct to date/time of sample*
- If the half-life is inaccurate, or ignores equilibrium in serial decay chains, significant bias may be introduced
- Have you checked your libraries' half-lives lately?

Half-Life: Example #1

An irradiated uranium PT sample was distributed for gamma ray fission product analysis. The following result was submitted:

Radionuclide	Library Half-Life, d	Activity, pCi/L	Uncertainty (k = 2)
^{140}Ba	12.75	$9.29x10^3$	$9.24x10^2$
^{140}La	1.68	$7.17x10^6$	$5.64x10^5$

Is this a realistic result? Why or why not?

Half-Life: Example #2

An irradiated uranium PT sample was distributed for gamma ray fission product analysis. The following result was submitted:

Radionuclide	Library Half-Life, d	Activity, pCi/L	Uncertainty (k = 2)
^{132}Te	3.2	1.50×10^{3}	1.0×10^{3}
^{132}I	0.095	9.5×10^{21}	1.6×10^{21}

?

Energies - Match the Library to the Sample

Possibilities for different types of libraries

- Uncontaminated environmental samples containing naturally occurring radionuclides
  - Soil, water and air samples require different assumptions regarding equilibria and half-lives.
- Samples contaminated with reactor waste
- Samples contaminated with fission event radionuclides
- Samples contaminated with wastes from industrial sources
- Client specific process samples or waste samples

Energy – The Qualitative Determinant

- Limits on gamma ray energy distinction/identification
  - Full width half maximum (FWHM) for the detector
  - Software algorithm
  - Selected presets on the software algorithm
- The energy variation for a single gamma ray energy on a QC control chart can be 0.2-0.3 keV or more, depending on the energy and the detector properties

Energy – The Qualitative Determinant (cont.)

- FWHM is a semi-quantitative measure of peak "goodness"

- The FWHM varies from ~ 0.6 keV at 60 keV to ~2.0 keV at 2,000keV (depending upon the detector)

Energy: Example #3

- The sample is counted 20 days post-event and…
- …the software identifies a peak at 667.45 keV
- …you have selected an energy tolerance/difference between measured and library value of 0.5 keV
- The peak has a 1 sigma counting uncertainty of about 2%
- There are lots of other gamma rays from other radionuclides present

Example #3: Parameters to Consider

The sample gamma ray is within 0.5 keV of the potential radionuclide candidates in the table below

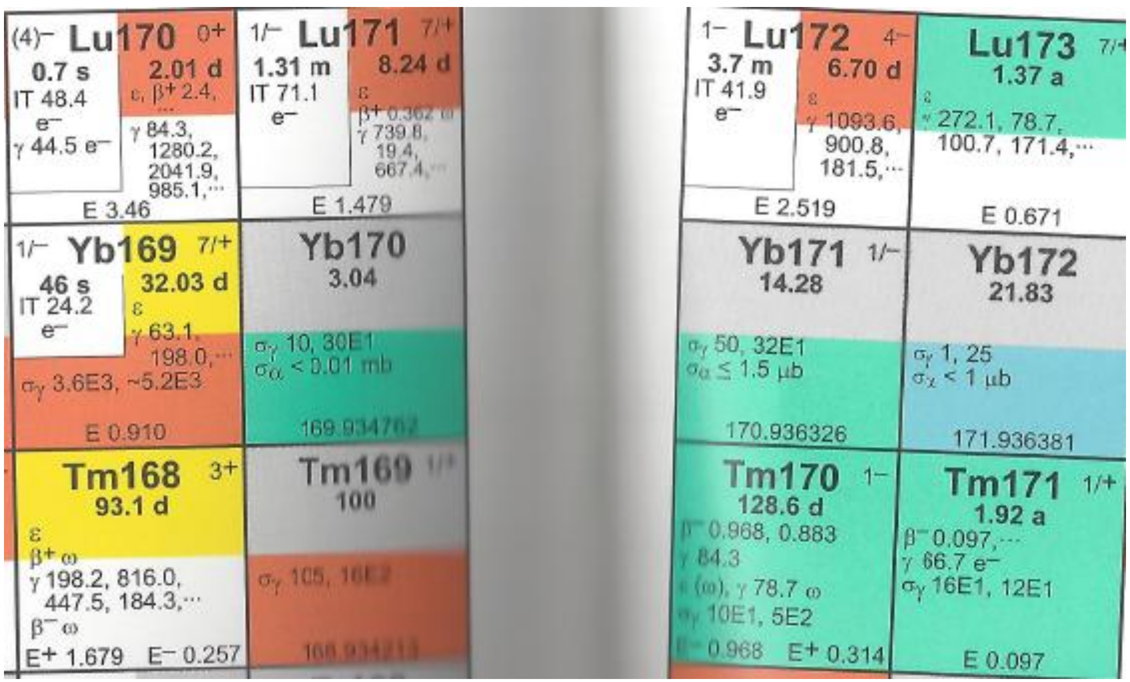
- Can they all be present and if so can they be distinguished?
- Which ones can be eliminated if a fission event occurred 4 days before the sample was taken?

Gamma ray Energy, keV	Radionuclide	Abundance, %	Half-Life, days
667.2	^{151}Er	16.7	$2.72x10^{-4}$
667.42	^{171}Lu	11.1	8.24
667.5	^{127}Sb	0.74	3.85
667.71	^{132m}I	13.9	$5.78x10^{-2}$
667.71	^{132}I	98.7	$9.5x10^{-2*}$

**When in equilibrium with parent ^{132}Te half life is 3.2 d*

Example #3: Eliminate Possibilities

- ^{171}Lu and ^{151}Er cannot be formed in a fission event
  - Wrong side of the line of stability
  - No stable isotope with (N-1) to activate



Example #3: Decision

- ^{132m}I has a direct fission yield of less than 0.01 %, is not fed from the potential isobaric progenitor ^{132}Te, and has a very short half life compared to 4 days
- The ratio of counts of ^{132}I to ^{127}Sb is ~160:1 taking into account their gamma ray abundances and half lives. Taking into account the difference in their mass chain fission yields the ratio exceeds 4,400
- Although ^{127}Sb is possible, its activity is not determinable from this energy peak due to the large counts from ^{132}I, AND it has gone through almost 6 half-lives

Energy: Example #4

- A PT sample that was spiked with fission and activation products was analyzed
- NORM not part of sample mix
- ^{214}Bi was reported as 4.53×10^{-7} µCi/mL
- Printout of the peak analysis activity report is on the next page

Nuclide Name	Id Confidence	Energy (keV)	Yield (%)	Activity (uCi/ml)	Activity Uncertainty
BI-214	0.517	76.86	0.45		
		79.29*	0.76	2.95894E-004	2.36366E-005
		89.80	0.34		
		273.70	0.17		
		387.00	0.29		
		389.10	0.37		
		405.74	0.18		
		454.77	0.28		
		469.69	0.14		
		609.31*	44.80	3.94054E-006	3.20244E-007
		665.45	1.29		
		719.86	0.42		
		768.36	4.80		
		786.10	0.30		
		806.17	1.12		
		934.06	3.03		
		964.08	0.38		
		1051.96	0.34		
		1069.96	0.28		
		1120.29*	14.80	5.17414E-007	1.18651E-007
BI-214	0.517	1133.66	0.28		
		1155.19	1.64		
		1207.68	0.49		
		1238.11	5.86		
		1280.96	1.44		
		1377.67	3.92		
		1385.31*	0.89	1.00222E-005	2.85794E-006
		1401.50*	1.55	1.46739E-005	3.65183E-006
		1407.98*	2.80	2.77374E-006	9.88748E-007
		1509.23	2.12		
		1538.50	0.51		
		1543.32	0.33		
		1583.22	0.70		
		1594.73	0.31		
		1599.31	0.38		
		1661.28	1.14		
		1683.99	0.25		
		1729.59	2.88		
		1764.49*	15.36	3.18320E-007	1.06641E-007
		1838.36	0.40		
		1847.42	2.04		
		1873.16	0.25		
		2118.55	1.14		
		2204.21	4.86		
		2293.36	0.30		
		2447.86	1.50		

^{131}I – peak abundance is 2.6% not in library

^{103}Ru (610.3 keV not in library)

Sum Peak 756 +364

^{110m}Ag – not in library

^{134}Cs sum peak

^{152}Eu (not in library)

FWHM was OK; Bi?

What went wrong…

- A NORM based library was used
- Only the most abundant gamma ray from the non-NORM radionuclides was in the library
- Activity concentrations ranged from 1.18×10^{-7} to 2.95×10^{-4} µCi/mL – (no review of the values)
- Weighted average was used, but gamma lines were eliminated and not shown on the report

Energy and Abundance Abuse: Example #5

- Gamma Ray Used Twice
- Gamma ray at 609-610 keV
  - ^{103}Ru, or
  - ^{214}Bi, or
  - Both

Energy and Abundance Abuse: Example #5 Libraries and Interference Corrections

Beware of duplicate energy lines in the library

- Irresolvable solutions may result when two radionuclides are attributed to one or more photopeaks within the match tolerance

- Interference correction may (or may not) help
  - Understand how your software works (capabilities and limitations)
  - Build libraries accordingly

- Thoroughly test interference corrections prior to use!

Example #5

Nuclide	Energy	Area	%Abn	%Eff	Uncorrected pCi/LITER	Decay Corr pCi/LITER	2-Sigm %Erro
NB-95	765.80	146501	99.81*	1.951E+00	7.532E+04	7.891E+04	9.69
ZR-95	724.19	37825	44.27	2.046E+00	4.181E+04	4.380E+04	10.47
	756.73	41470	54.38*	1.971E+00	3.874E+04	4.058E+04	10.56
RU-103	497.08	70512	91.00*	2.805E+00	2.765E+04	2.983E+04	14.41
	610.33	3769	5.76	2.364E+00	2.771E+04	2.989E+04	18.90
AG-110M	657.76	348	94.30*	2.219E+00	1.664E+02	1.684E+02	55.95
	677.62	------	10.56	2.163E+00	------	Line Not Found	------
	706.68	------	16.33	2.087E+00	------	Line Not Found	------
	763.94	146501	22.62	1.951E+00	3.323E+05	3.363E+05	9.90
	884.68	325	72.70	1.724E+00	2.593E+02	2.624E+02	24.13
	937.49	171	34.20	1.642E+00	3.047E+02	3.084E+02	31.91
	1384.29	80	24.90	1.191E+00	2.697E+02	2.730E+02	29.99
I-131	80.19	9575	2.62	4.840E+00	7.558E+04	1.095E+05	10.86
	284.31	22065	6.12	4.348E+00	8.300E+04	1.203E+05	9.60
	364.49	255240	81.50*	3.603E+00	8.700E+04	1.261E+05	9.10
	636.99	14510	7.16	2.280E+00	8.897E+04	1.289E+05	10.43
CS-134	563.25	821	8.34	2.528E+00	3.898E+03	3.914E+03	42.94
	569.33	1480	15.37	2.505E+00	3.847E+03	3.862E+03	26.22
	604.72	8358	97.62	2.382E+00	3.597E+03	3.612E+03	10.67
	795.86	5934	76.91*	1.888E+00	4.091E+03	4.108E+03	10.00
	801.95	601	8.69	1.875E+00	3.693E+03	3.707E+03	14.27
	1365.19	200	3.02	1.204E+00	5.501E+03	5.523E+03	18.74
BA-137M	661.66	375	89.90*	2.208E+00	1.891E+02	1.891E+02	41.71
CS-137	661.66	375	85.10*	2.208E+00	1.997E+02	1.998E+02	41.72
CE-139	165.86	9832	80.00*	5.944E+00	2.070E+03	2.115E+03	10.86
CE-141	145.44	190044	48.29*	6.175E+00	6.379E+04	6.991E+04	8.36
BI-214	609.32	3769	45.49*	2.364E+00	3.509E+03	3.509E+03	13.86
	1120.29	24	14.92	1.413E+00	1.143E+02	1.143E+02	144.83
	1764.49	------	15.30	9.984E-01	------	Line Not Found	------

Note the abundance ratios make the ^{214}Bi improbable!

Example #6: X-ray/Energy Confusion

- X-rays come from electronic transitions outside the nucleus. If a radionuclide
  - decays by EC or positron emission, the x-rays are those of the progeny nuclide
  - decays by IC, the x-rays come from the same radionuclide as would have emitted the gamma ray
- The x-ray energies for an atom are independent of A and only dependent upon Z
  - If an x-ray is used for analysis of a specific radioisotope *no other radioisotope of that element* may be present
  - X-rays are generally considered to be interferences in gamma ray spectrometry

Example #6

^{131}I (major), ^{144}Ce (minor)

pk	energy	area	uncert	fwhm	nuclide	brnch.	act.	nuc
	80.14	5225.	4.44	1.05	80.12	1.600	1.785E+04	CE144

Nuclide	Peak Channel	Centroid Energy	Background Counts	Net Area Counts	Intensity Cts/Sec	Uncert 1 Sigma %	FWHM keV
J-129	117.10	29.46	13792.	1069.	0.099	15.90	0.851
J-129	118.38	29.78	12739.	3507.	0.325	3.45	0.851
Ba-133	131.90	33.16	28434.	4517.	0.418	5.48	0.855
CE-139	131.37	33.03	18768.	1910.	0.177	7.65	0.855
CE-139	133.00	33.44	21828.	3199.	0.296	6.38	0.855
J-129	133.64	33.60	18507.	719.	0.067	22.44	0.856
XE-131M	136.84	34.40	45389.	-3972.	-0.368	7.76	0.856
J-129	136.84	34.40	18261.	6797.	0.629	3.06	0.856
CE-141	141.43	35.55	32984.	11498.	1.065	6.50	0.858
CE-141	143.34	36.03	26530.	19944.	1.847	1.04	0.858
J-131	1134.32	283.99	10890.	8333.	0.772	2.92	1.263
EU-152	1372.28	343.52	9191.	411.	0.038	32.81	1.173
J-131	1454.80	364.16	8473.	99871.	9.247	0.38	1.283

X-rays: not specific to ^{129}I
(^{131}I has IC/γ = 1.2 at 80 keV and 0.05 at 283 keV

X-rays: not specific to ^{141}Ce

Unidentified Gamma Rays: Example #7

- A PT sample with several spiked radionuclides (fission and activation products)
- Six gamma ray energies not identified
- Peak areas were comparable to other gamma rays in the spectrum

Example #7

Unidentified Energy Lines
Sample ID : x101116011001

Pb X-ray
^{139}Ce
^{131}I
^{110m}Ag

Energy	Area	Bkgnd	FWHM	Channel	Left	Pw	Cts/Sec	%Err	%Eff
74.87	271	5569	1.10	148.46	147	6	2.51E-02	91.8	1.81E+00
165.98	2774	10650	1.16	330.87	327	9	2.57E-01	13.9	4.02E+00
325.36	389	5258	1.24	649.99	646	9	3.60E-02	67.8	3.05E+00
503.06	349	2268	1.60	1005.79	1002	8	3.23E-02	48.7	2.39E+00
657.59	183	1067	1.70	1315.19	1311	9	1.69E-02	66.1	2.03E+00
884.77	133	334	1.96	1770.06	1764	11	1.23E-02	56.0	1.67E+00

Gamma Ray Abundances

- Have you checked your listings against those used by your standards supplier?
  - Do they agree within the stated uncertainty?
  - Is the uncertainty and the number of significant figures sufficient to achieve the number of significant figures in reported results?

Document and Protect Your Libraries

- Keep careful control of your libraries!
- Document the source(s) of nuclear data and deviations from published nuclear data along with the rationale for changes
- Document “on-the-fly” modifications to library
- Beware of library “creep” - protect library files from being overwritten with modified library files

Practice, Practice, Practice…

- The EPA NAREL Mixed Fission PT program using different gamma radionuclides
  - as spiked solutions and
  - as irradiated U

  is providing valuable experience for participants and the program

- The greater the variety of radionuclide mixes that are used, the more experience the labs will get, and the better will be our confidence in future analytical results

Activity Calculations and Spectrum Review

Activity

$$A_E = \frac{N_E}{\varepsilon_E \times t \times \gamma_E \times \mathrm{f} \times e^{-\lambda T}}$$

Where:

A_E = *the activity of the nuclide based on energy E*

N_E = *the net peak area for the peak at energy E*

ε_E = *the detector efficiency at energy E*

t = *the live time for the sample count*

γ_E = *the gamma intensity of this nuclide at energy E*

λ = *the decay constant of this nuclide*

T = *the elapsed time between the reference date and the count*

f = *other factor(s) which may be applicable*

Activity Concentration

$$AC_E = \frac{A_E}{\text{V}}$$

Where:

AC_E = the activity of the nuclide based on energy E

A_E = the activity of the nuclide based on energy E

υ = the mass or volume of the sample or aliquot

Abundance Weighted Mean Activity (or Activity Concentration)

$$\mathrm{AC} = \frac{\sum_{i}^{n}(AC_i \times \gamma_i)}{\sum_{1}^{n}\gamma_i}$$

Where:

AC = the abundance weighted mean activity (or activity concentration) of the nuclide

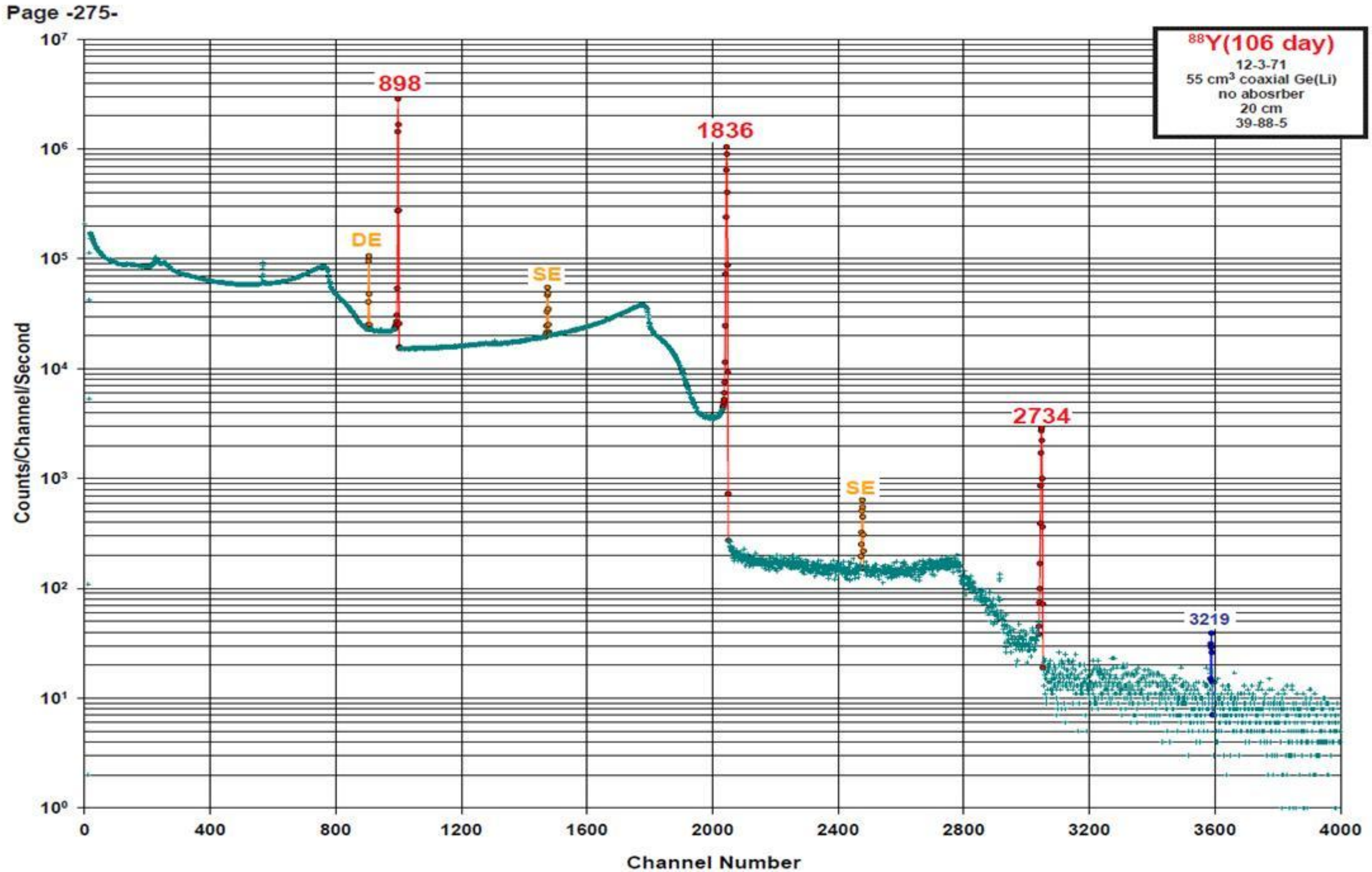
γ_E = the gamma abundance for this nuclide at energy E

n = *the number of gamma rays used in the analysis*

Spectrum Interpretation – Simple Spectra

1. Clean, separated peaks
2. No interferences or deconvolution
3. Simple peak fitting
4. Simple peak area calculation

Example Spectrum from Y-88 Decay



Spectrum Interpretation – Complex Spectra

1. Overlapping or very close peaks
2. Interference correction and/or deconvolution necessary
3. Complex peak fitting – close review necessary
4. Complicated peak area calculation

Energy (keV)	ROI start	ROI end	Peak Centroid	Net Peak Area	Net Area Uncertainty	Continuum Counts	FWHM (keV)
176.39	349 -	359	353.02	1.80E+03	137.73	2.05E+03	1.60
380.60	757 -	768	761.45	2.14E+02	72.43	8.72E+02	1.45
428.11	849 -	863	856.47	4.26E+03	150.54	9.77E+02	1.92
463.57	919 -	934	927.38	1.40E+03	98.21	6.73E+02	1.52
510.67	1016 -	1029	1021.59	6.89E+01	45.85	3.30E+02	1.19
600.88	1194 -	1221	1201.99	1.93E+03	82.08	2.24E+02	1.88
606.81	1194 -	1221	1213.85	6.33E+02	47.20	2.29E+02	1.88
636.22	1264 -	1279	1272.65	1.20E+03	75.18	1.38E+02	1.88
662.00	1316 -	1350	1324.22	5.45E+02	45.02	1.34E+02	1.82
671.84	1316 -	1350	1343.89	1.93E+02	27.14	1.20E+02	1.82
796.17	1585 -	1599	1592.52	1.46E+02	31.24	6.78E+01	1.70
810.88	1614 -	1628	1621.94	7.47E+01	27.40	7.87E+01	1.59

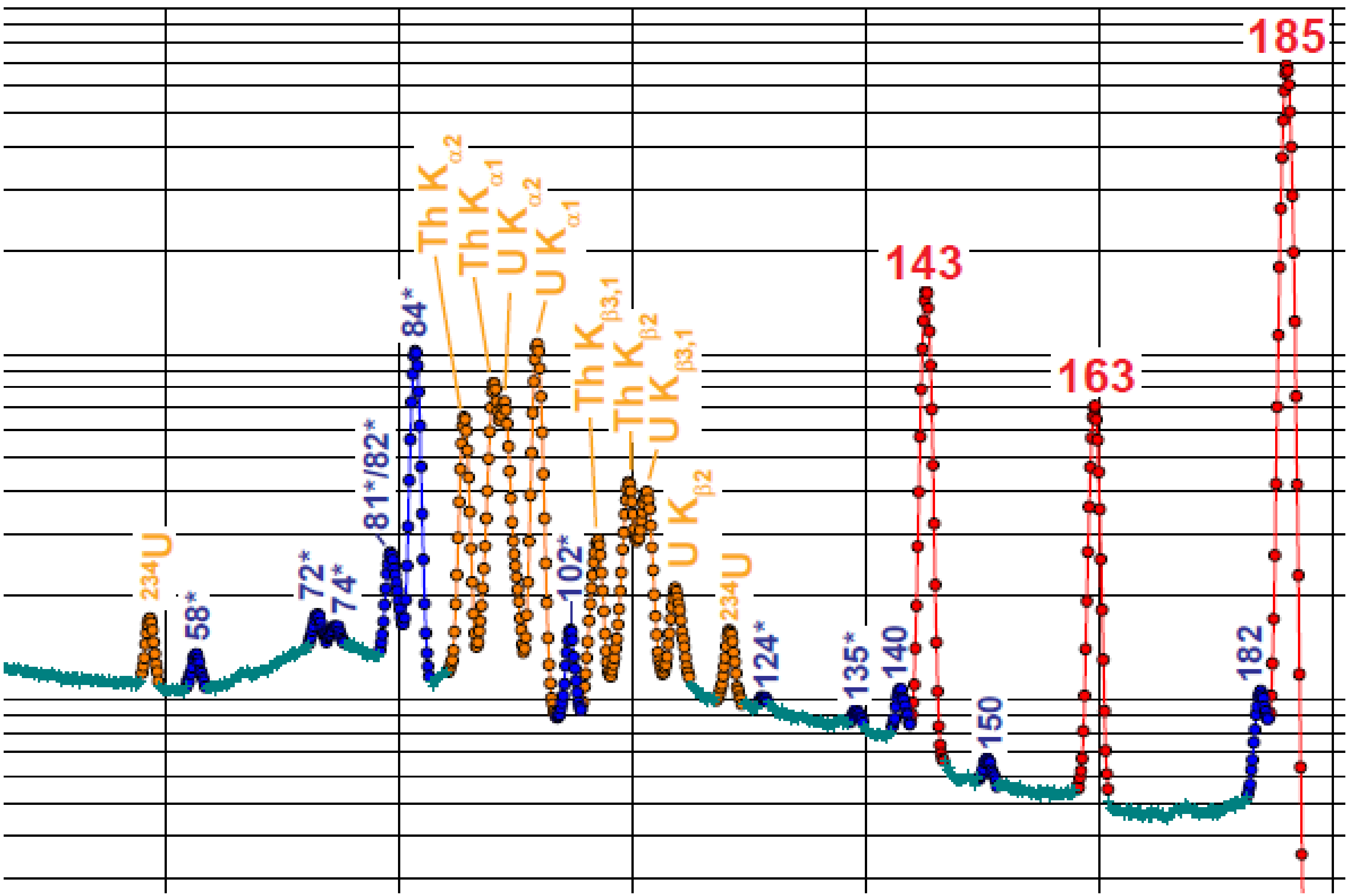
UNIDENTIFIED PEAKS

Peak Locate Performed on : 8/8/2012 1:15:15AM
Peak Locate From Channel : 115
Peak Locate To Channel : 4096

Peak No.	Energy (keV)		Peak Rate (CPS)	Peak Rate (%) Uncertainty
5	510.67	Ti-208	1.72E-02	33.27
11	796.17	Pa 234	3.65E-02	10.69
15	1461.47	K-40	9.42E-03	18.22

Gamma printout from a NPP liquid effluent release

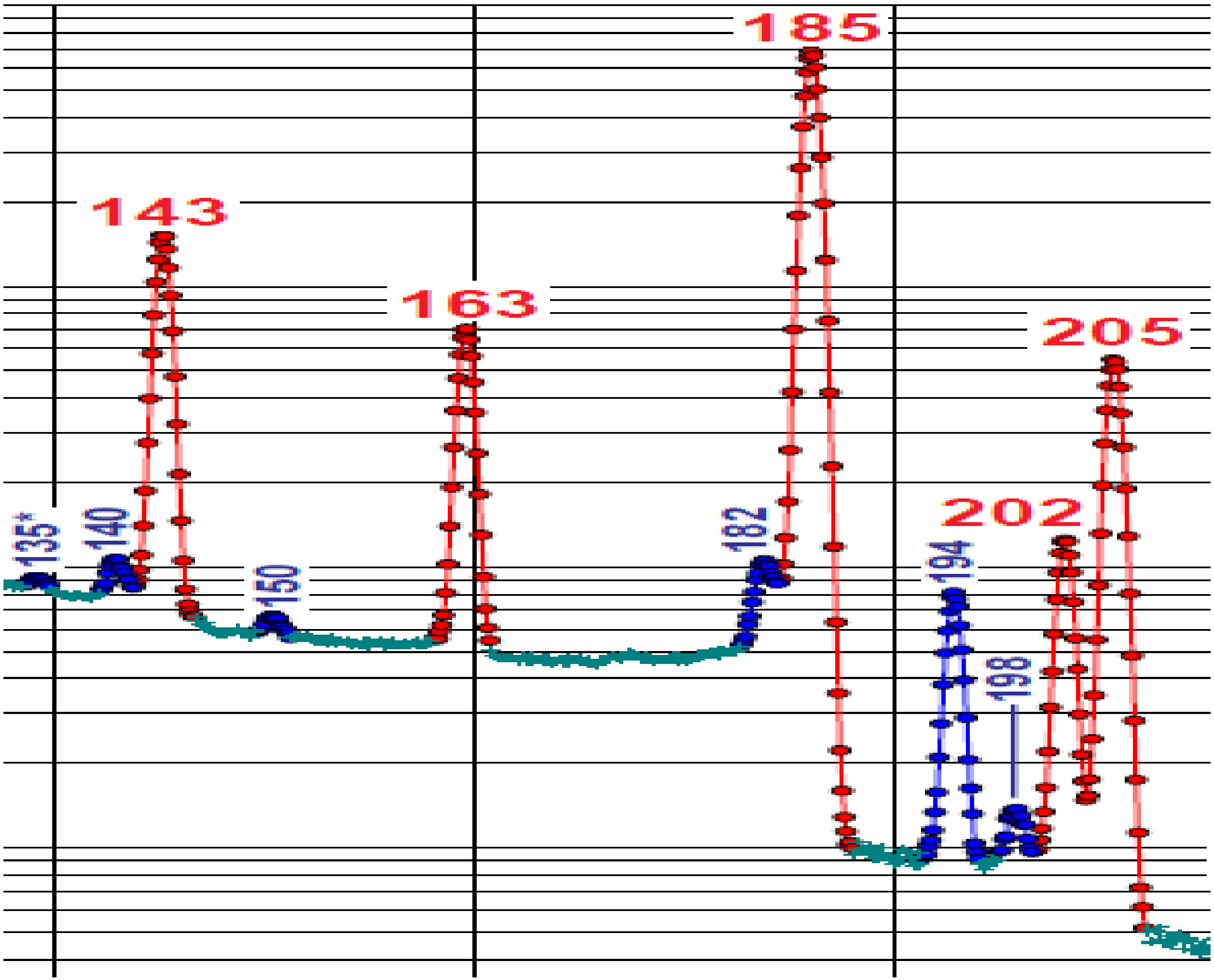
Spectrum Interpretation – Complex Spectrum - SNM



Peak Stripping (Interference Correction)

- Sources of interference
- Consequences of not correcting
- Basic principles of interference correction
- Example interference correction

Interference Correction Example



Interference Correction Example

Because ^{235}U and ^{226}Ra both contribute to the peak at 186 keV, we must calculate the activity of ^{235}U present in the sample using line(s) without interferences

At 143.8 keV (for example)

1419 cts in 2400 sec

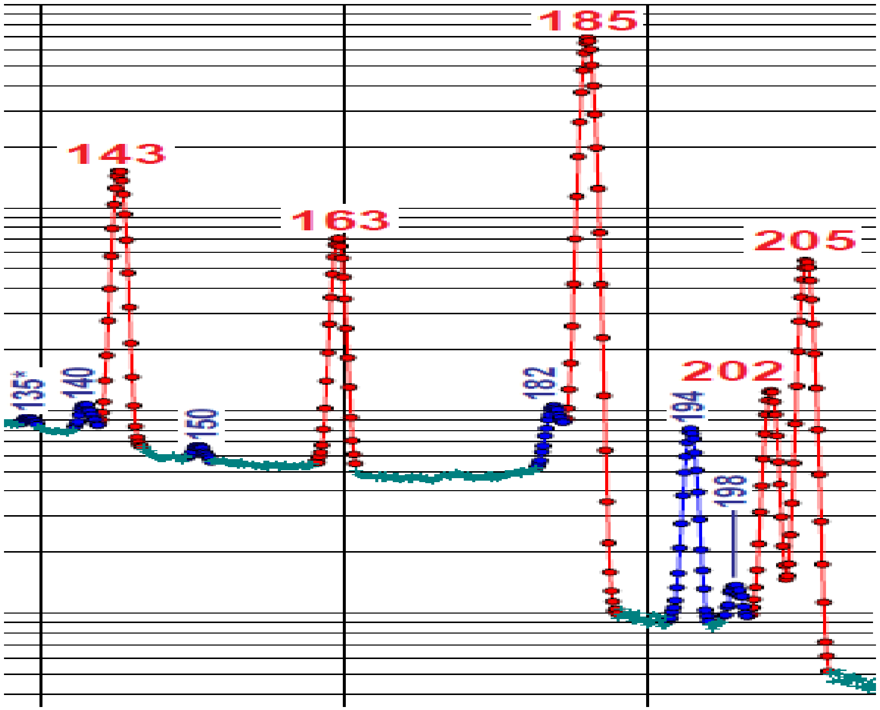
Eff = 0.069 cps/gps (gps=gamma/sec)

BR = 0.1053 gamma/dis

Act_{235U} = Cts / t / Eff / BR

gps = 1419 / 2400 / 0.069 / 0.1053

Act_{235U} = 81.38 dps (Bq) ^{235}U



Interference Correction Example

Knowing the ^{235}U activity in the sample, we can calculate the number of ^{235}U counts present (interfering) with the 186 keV peak

^{235}U activity = 81.38 dps

At 186 kev

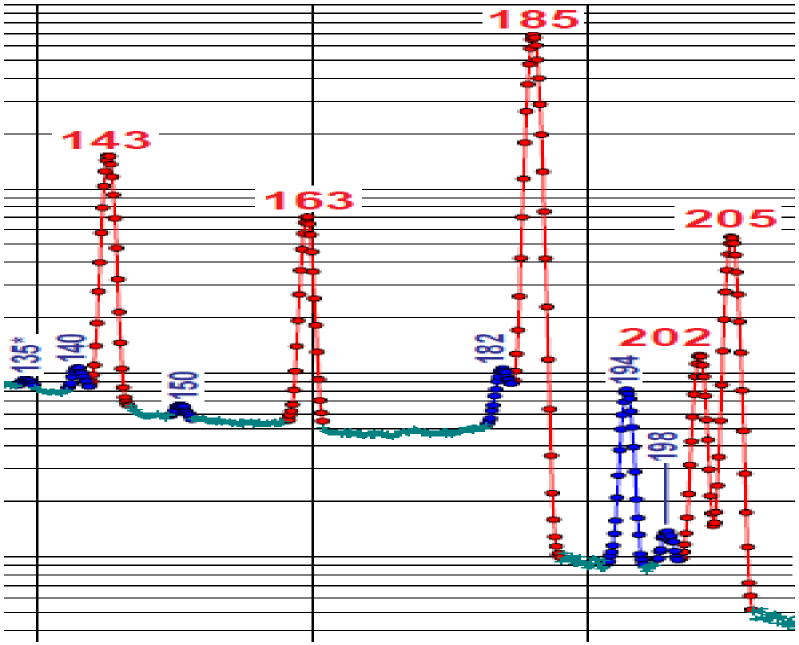
Eff = 0.0716 cps/gps

BR = 0.5315

$Cts_{235U\text{-}186keV} = Act_{235U} * t * Eff * BR_{235U\text{-}186keV}$

$= 81.38 * 2400 * 0.0716 * 0.5315$

$= 7432$ cts ^{235}U



Interference Correction Example

The difference between the observed counts in the 186 keV peak and the calculated ^{235}U counts is attributable to ^{226}Ra. Now we can calculate how much ^{226}Ra is present.

$Cts_{Total\text{-}186keV}$ = 8174 cts $Cts_{235U\text{-}186keV}$ = 7432 cts

$Cts_{226Ra\text{-}186keV}$ = 8174 cts - 7432 cts

= 741.7 cts ^{226}Ra

@186 keV: Eff =0.0716, BR = 0.0328

Cnt time = 2400 sec so:

Act_{226Ra} = 742 / 2400 / 0.0716 / 0.0328

Act_{226Ra} = 132 dps (Bq)

Summing in Gamma Spectrometry

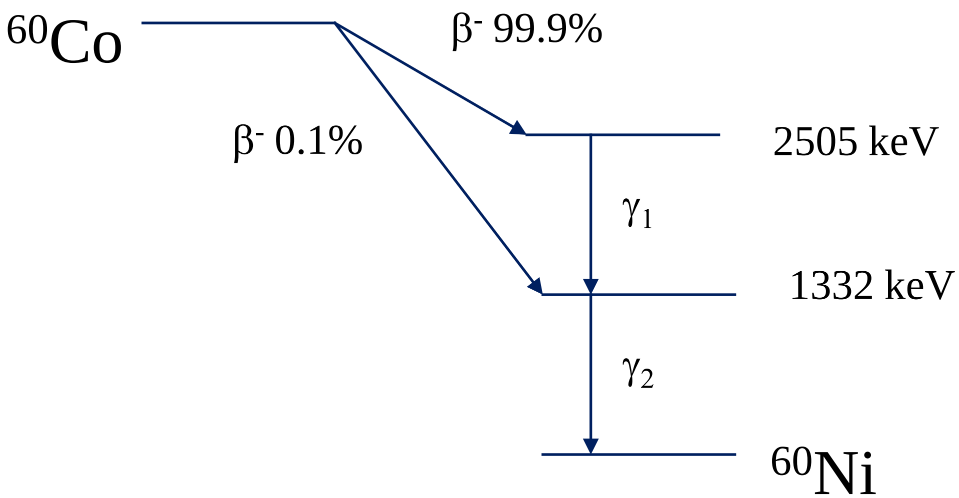
Summing in General

- Two or more photons strike detector at the same time (i.e., within detection system resolving time)
  - These counts disappear from where they would normally be found in the spectrum (they sum out)
  - A count is gained at the combined energy of the absorbed photons (it sums in)
    - If counts are lost from two FEPs the resulting peak would have the combined energy of the FEPs
    - Sample counts lost → low bias to sample result
    - Standard counts lost → efficiency biases low, sample high

The Decay Scheme Influences Spectral Features

- ***Coincidence*** describes the nearly "simultaneous" emission of two or more photons from the same decay event
  - Examples include:
    - The gamma rays and X-rays that follow electron capture
    - The annihilation photons and gamma rays that follow positron emission
    - Two photons emitted in cascade (sequentially)
- How can you tell when two gammas are in coincidence?
  - Review the level scheme for that radionuclide

Coincident Photons



$$\gamma_1 = (2505.7 - 1332.5) = 1173.2 \text{ keV}$$

$$\gamma_2 = (1332.5 - 0) = 1332.5 \text{ keV}$$

Coincident photons may be emitted by nuclei that have decay schemes that have one transition following another (in cascade) as shown above

True Coincidence Summing (TCS)

- Two or more coincident photons are emitted within the resolving time of the detection system
  - The photons interact with and deposit their full energy in the detector.
  - The photons are not resolved separately, rather a single pulse is processed with amplitude corresponding to the combined energies of the individual photons.
- A single photopeak may be observed at the combined energy of the two individual FEPs

True Coincidence Summing (TCS) (cont.)

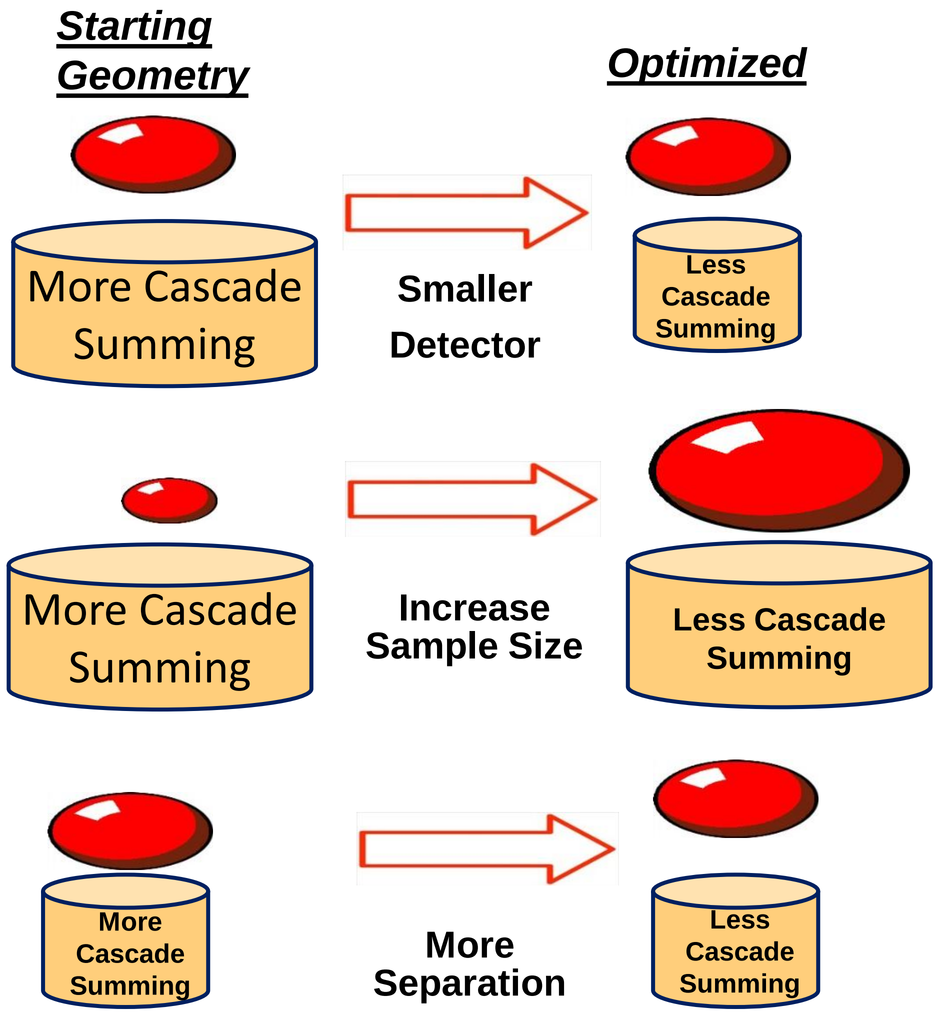
- Coincidence summing is independent of count rate.
  - The probability that two coincident gamma rays will sum with one another is proportional to the chance they will both interact with, and deposit their full energy, in the detector.
  - Gamma and x-rays with energies that span the "knee" of a detector efficiency curve are more likely to sum

Can True Coincidence Summing be Minimized?

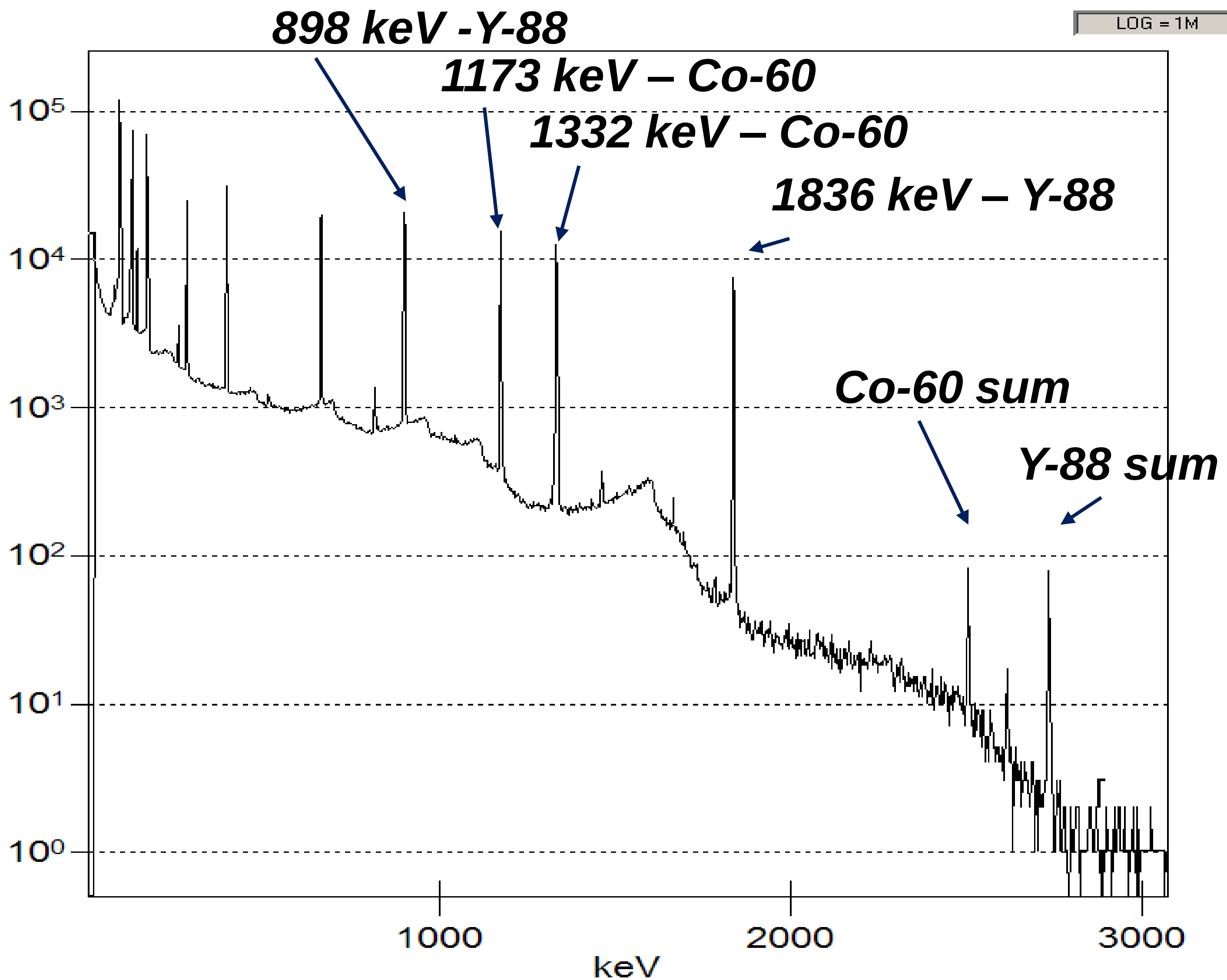
- TCS is most severe with
  - Radionuclides with complex decay schemes
  - Geometries close to detector (e.g., air filters on endcap)
  - Larger detectors and well detectors
  - Detectors sensitive to low-energy photons (x-rays)
- TCS is not affected by activity level or count rate
- TCS can be reduced by decreasing the efficiency
  - Summing is proportional to the product of the detection efficiencies of the nuclides doing the summing.

True Coincidence Summing Geometry Dependencies

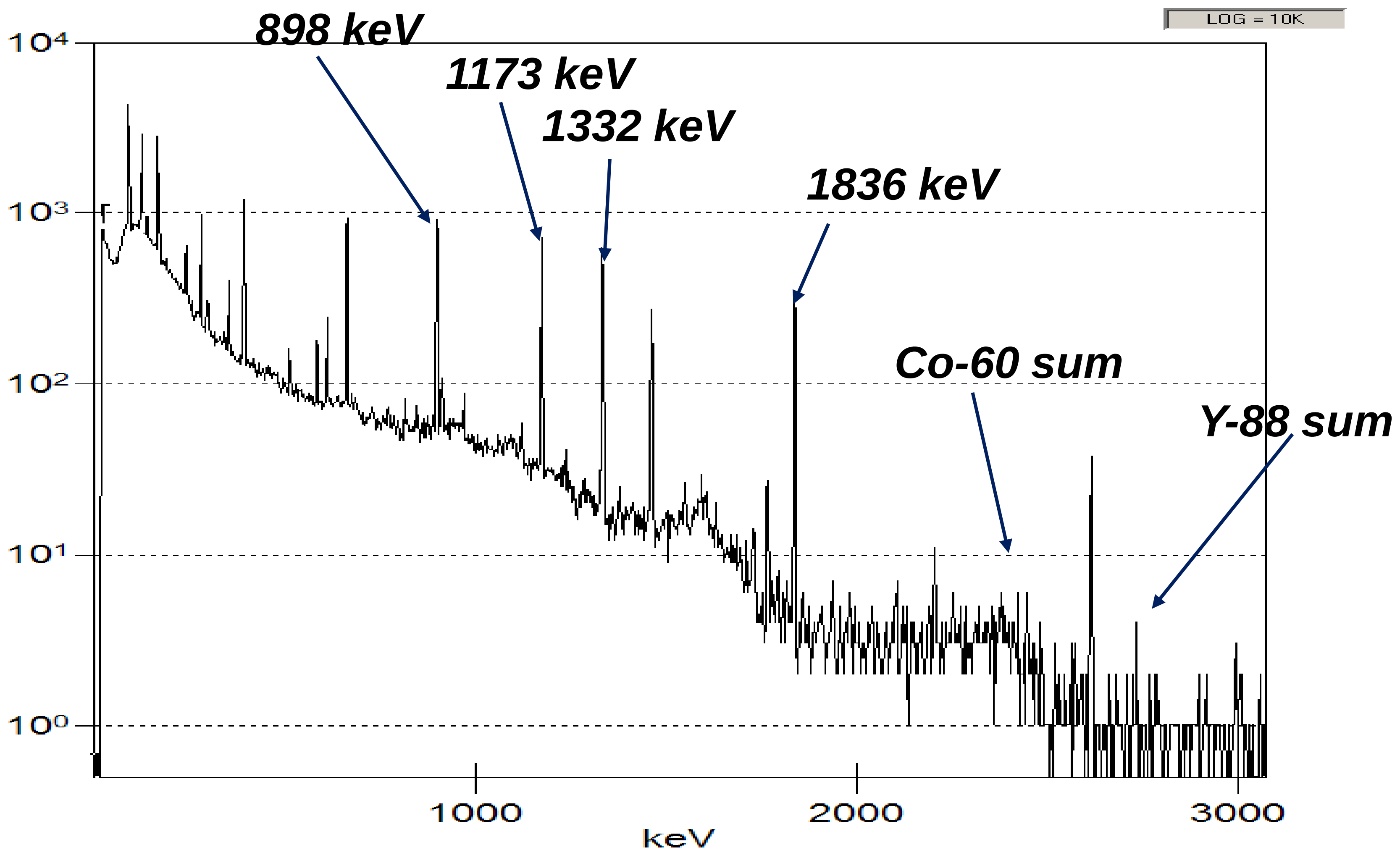
- *Detector Size*
- *Sample Size (Dilution)*
- *Separation (Standoff)*



Spectrum from Mixed Gamma Source Unshielded on *Face of Detector*



Spectrum from Mixed Gamma Source Unshielded *10 cm from Detector*



True Coincidence Summing

- Higher energy levels in a cascade emit real gamma rays
  - Gammas from the cascade below may sum to this energy as well!
- A Compton edge is not observed below pure sum peaks
  - A Compton edge may be observed below a combined sum and gamma peak, however, if the emission abundance direct from the summed energy level state is significant (i.e., 2505 keV gamma from 60Co)

Na-22 Example

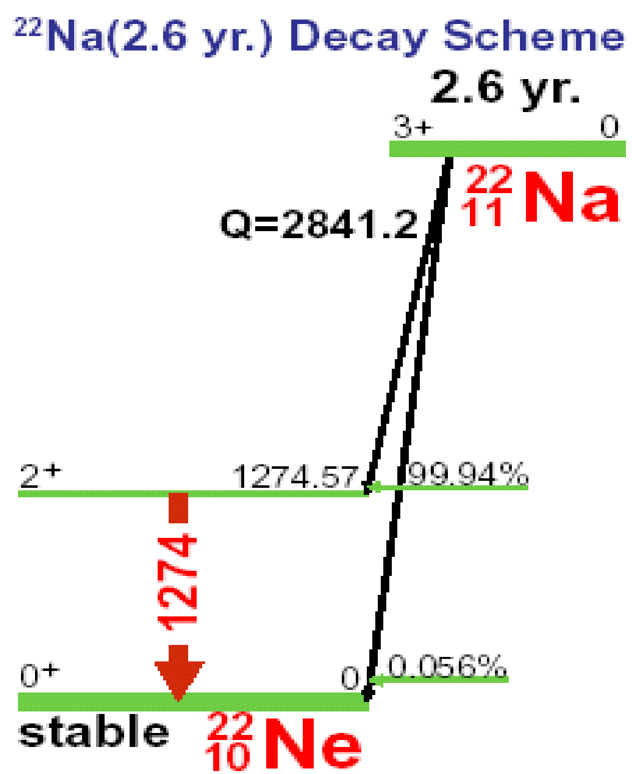
GAMMA-RAY ENERGIES AND INTENSITIES

Nuclide: ^{22}Na — Half Life: 2.6019(4) yr.

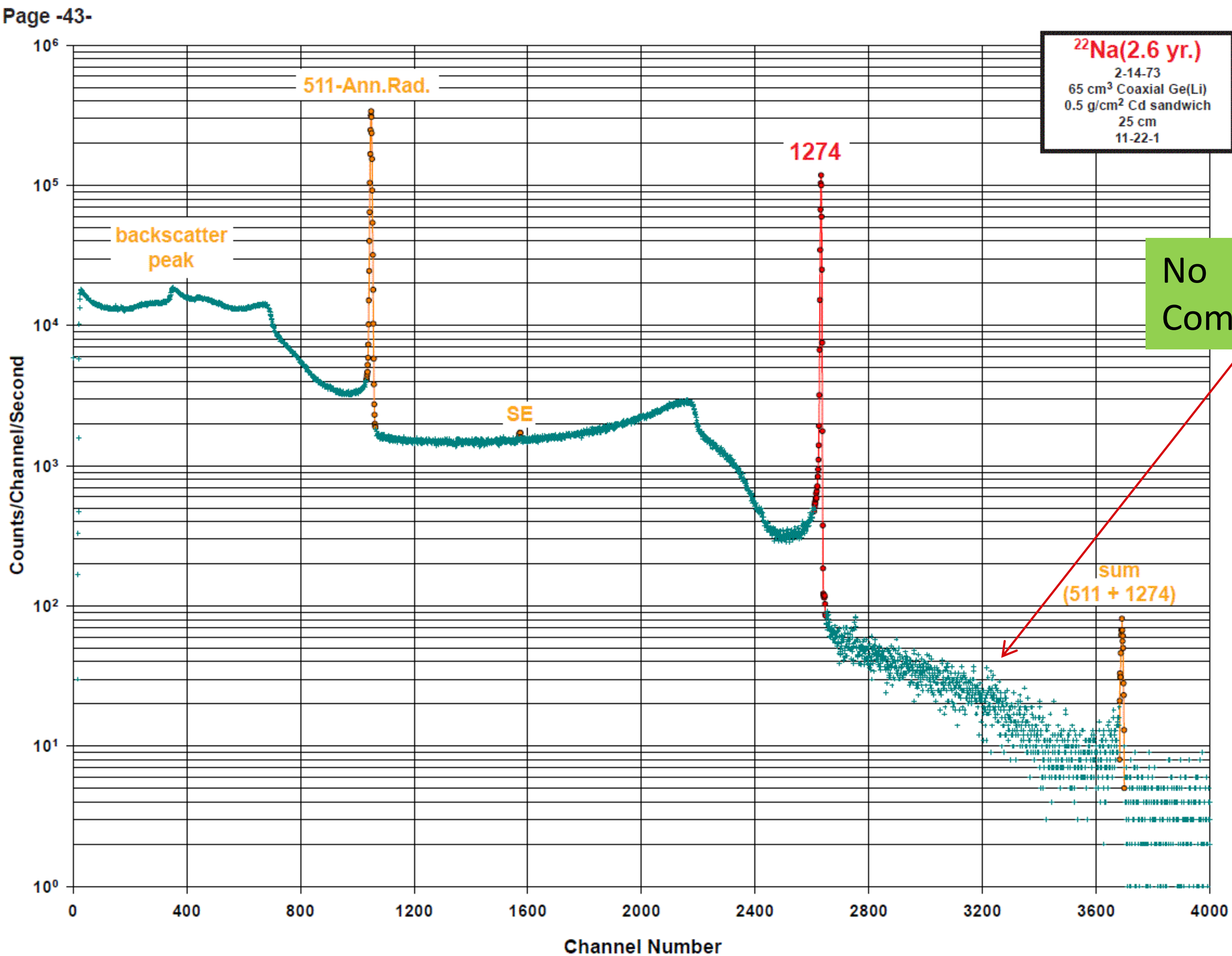
Detector: 55 cm³ coaxial Ge (Li) — Method of Production: Ne(^{3}He,p)

	E_γ (keV)	σE_γ	I_γ (rel)	I_γ (%)	σI_γ	S
Ann.	511.006		100	178.0	0.6	1
	1274.53	0.02	62.2	99.944	0.014	1

E_γ, σE_γ, I_γ, σI_γ - 1998 ENSDF Data



Na-22 Example cont'd



No Compton

Radionuclides with True Coincidence Sum Effects

Radionuclide	Gamma 1, keV	Gamma 2, keV	Sum Peak, keV
^{60}Co	1173	1332.5	2505*
^{134}Cs	604	796	1400
^{154}Eu	58.4	1274	1332.8
	123.1	1047.2	1170.3
^{88}Y	1836	898	2734**

*Non-coincidence abundance is 2.0×10^{-6} %

**Non-coincidence abundance is 7.10×10^{-1} %

Corrections for True Coincidence Summing

- Software / computer code / calculational models
  - Gamma spectrometry software packages have provisions for TCS correction although they require that the detector be characterized and/or calibrated with special sources and a true coincidence calibration method applied
  - Not recommended unless the approach is thoroughly understood, work is done to fine tune models, and the procedure is validated prior to use

Corrections for True Coincidence Summing

- Best (i.e., most accurate) "fix" involves leveraging the principles we have discussed
  - Calibrate the detector with the radionuclide of interest in the geometry in which samples will be counted
  - This is the most accurate technique because summing in the sample and the calibration standard are identical
- Easiest "fix"
  - Move sample to a calibrated position further from the detector

Random Summing (Pulse Pile-Up)

- High source activity results in count rates that exceed the resolving capability of detector
  - **Strictly** a function of count rate
  - A random sum peak may be observed at the combined energy of the highest count rate peaks
- Can be reduced by decreasing the count rate:
  - Take less activity by decreasing size of the aliquant
  - Increase distance between the center of the sample and the detector

Random Sum Events from PT Study

Radionuclide	Photon1, keV	Photon 2, keV	Sum Peak, keV	Sample Type
^{137}Cs	661	511	1173	Any high Activity ^{137}Cs
^{58}Co	810	511	1321	Reactor Coolant
	810	810	1620	Reactor Coolant
^{132}I + ^{140}Ba	667	538	1205	Fresh Fission Products
^{131}I + ^{239}Np	80	106 (X-ray)	186	Fresh Fission Products

Random Summing Correction

- Can be done empirically for idealized situations
  - Not recommended
- Best “fix”
  - Move sample to a calibrated position further from the detector
- Next best “fix”
  - Dilute sample

Gamma Spectrometry References

1. G. Gilmore, "Practical Gamma Ray Spectrometry," 2nd Edition, John Wiley and Sons (2008)
2. G. Friedlander, J. W. Kennedy, E. S. Macias and J. M. Miller, "Nuclear and Radiochemistry", John Wiley and Sons (1981).
3. G. Knoll, "Radiation Detection and Measurement", John Wiley and Sons (1979)
4. Multi Agency Radiological Laboratory Analytical Protocols (MARLAP) Manual, NUREG-1576, EPA 402-B-04-001A (July 2004)
5. ANSI N42.14, American National Standard for Calibration and Use of Germanium Spectrometers for the Measurement of Gamma-Ray Emission Rates of Radionuclides
6. ASTM D3649, Standard Test Method for High-Resolution Gamma-Ray Spectrometry of Water
7. ASTM D7282, Standard Practice for Set-up, Calibration, and Quality Control of Instruments Used for Radioactivity Measurements.
8. Interactive Chart of the Nuclides, http://www.nndc.bnl.gov/chart/
9. NUDAT Decay Radiation Data look-up, http://www.nndc.bnl.gov/nudat2/indx_dec.jsp
10. Le Laboratoire National Henri Becquerel, Table of Radionuclides, Recommended Data http://www.nucleide.org/DDEP_WG/DDEPdata.htm
11. Gamma Ray Spectrometry Catalogshttp://www.inl.gov/gammaray/catalogs/catalogs.shtml

Questions?

Future NAMP Radiochemistry Webinars

- Overview of EPA Rapid Methods (October 24)
- Subsampling (November 14)
- Mass Spectrometry (December 12)
- Guide to Uncertainty Measurement (January 23)
- Visit NAMP website at www.wipp.energy.gov/namp